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PAPER

Spin reversal in Fe₈ under fast pulsed magnetic fieldsS Narayana Jammalamadaka^{1,2}, S Véléz^{3,4}, J Vanacken¹, V V Moshchalkov¹, L F Chibotaru⁵, J Tejada³ and F Macià³¹ Pulsed Field Group, Institute for Nanoscale Physics and Chemistry (INPAC), KU Leuven, Celestijnenlaan 200D, B-3001 Leuven, Belgium² Department of Physics, Indian Institute of Technology Hyderabad, Hyderabad 502205, India³ Grup de Magnetisme, Departament de Física Fonamental, Universitat de Barcelona, Barcelona 08028, Spain⁴ CIC nanoGUNE, 20018 Donostia-San Sebastian, Basque Country, Spain⁵ Theory of Nanomaterials Group, Katholieke Universiteit Leuven, Celestijnenlaan 200F, B-3001 Leuven, Belgium**Keywords:** low dimension magnetism, spin dynamics, magnetic deflagration

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Abstract

We report measurements on magnetization reversal in the Fe₈ molecular magnet using fast pulsed magnetic fields of 1.5 kT s⁻¹ and in the temperature range of 0.6–4.1 K. We observe and analyze the temperature dependence of the reversal process, which involves in some cases several resonances. Our experiments allow observation of resonant quantum tunneling of magnetization up to a temperature of ~4 K. We also observe shifts in the maxima of the relaxation within each resonance field with temperature that suggest the emergence of a thermal instability—a combination of spin reversal and self-heating that may result in a magnetic deflagration process. The results are mainly understood in the framework of thermally-activated quantum tunneling transitions in combination with emergence of a thermal instability.

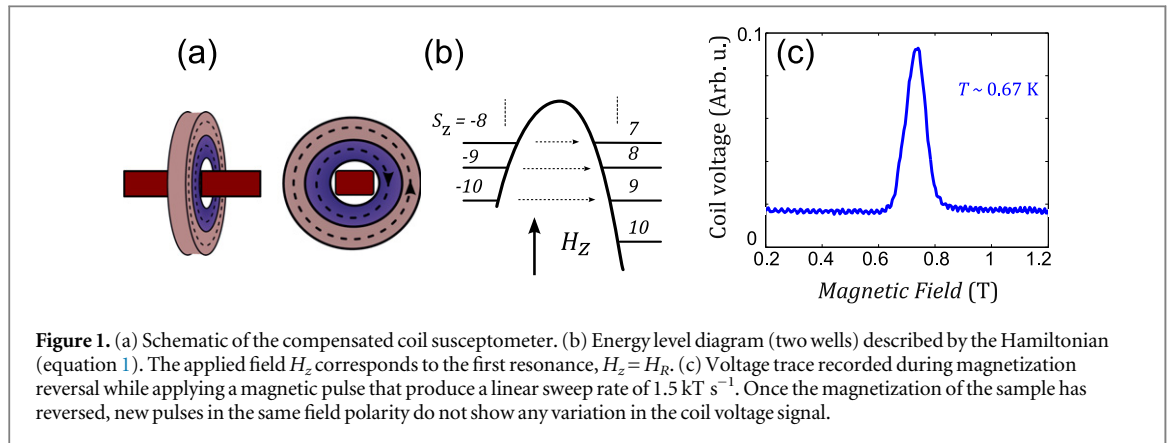
1. Introduction

Single molecule nanomagnets have been of great interest because of their quantum effects [1–7] and possible applications in quantum computers [8, 9] or magnetic refrigerants [10]. Spin-level populations in nanomagnets can be manipulated easily by modifying their energy with external magnetic fields. Among the large set of synthesized molecule magnets, Mn₁₂ and Fe₈ have been the most studied because of their relatively easy preparation, large molecular spin, and large magnetic anisotropy. Dynamics of spin at low temperatures had been studied—including quantum tunneling magnetization (QTM) [2] and electron paramagnetic resonance (EPR) [11, 12]—and has been described through the so-called giant spin approximation [1] that assigns a single spin quantum number, *S*, to the ground-state spin levels. Effectively, the spins relax toward equilibrium through a combination of thermal activation and quantum tunneling [2, 6].

Fe₈ was initially prepared by Wieghardt *et al* [13] and has shown clear evidence of QTM [5, 7, 14–16]. At low temperatures, the eight iron cations assemble couple in such a manner that give rise to a high spin *S* = 10 molecule with an anisotropy barrier height of about 29 K. The spin Hamiltonian for the Fe₈ is given by [14, 17]

$$\mathcal{H} = -DS_z^2 + E(S_x^2 - S_y^2) + \mathcal{H}_{\text{ho}} - g\mu_B \mathbf{H} \cdot \mathbf{S}, \quad (1)$$

where anisotropy constants *D* and *E* are 0.27 and 0.029 K respectively and *g* ≈ 2. Both anisotropies have been extensively measured through high frequency-EPR [17] and neutron spectroscopy [18]. The first term in the Hamiltonian defines the anisotropy barrier and creates an easy axis (*z*-axis) for the magnetization. The second and third terms break the rotational symmetry of the Hamiltonian and are responsible for the tunneling of the magnetization. The term \mathcal{H}_{ho} stands for high order terms. The last term of the Hamiltonian describes the Zeeman energy associated with an external applied field **H**. An applied field in the direction of the easy axis, *H_z*, biases the potential well reducing the activation energy and increasing the relaxation rate, particularly at specific resonant values of the magnetic field (*H_z* = *nH_R* for integer *n*) corresponding to the crossings of energy levels with opposite spin projections (see, figure 1(b)). Transverse fields *H_⊥* reduce the activation energy and mix eigenstates of *S_z* increasing relaxation of magnetization by quantum tunneling. Notice that a small or a moderate transverse field, *H_⊥* << 2*DS*/(*gμ_B*), does not affect the resonance values *nH_R*.



In this letter we investigate the temperature dependence of the magnetization reversal in the Fe_8 molecular magnet under fast pulsed magnetic fields of 1.5 kT s^{-1} . This fast sweep rate allows observation of quantum tunneling of the magnetic moment at high temperatures up to $\sim 4 \text{ K}$. The measurements of spin dynamics are modeled with thermally-activated quantum tunneling transitions accounting for the self-heating that might result in a magnetic deflagration [19].

2. Experimental set-up

A single crystal of Fe_8 molecular magnet with dimensions approximately $1 \times 0.15 \times 0.27 \text{ mm}$ was used for the present investigation. The sample was embedded in stycast, which was pored in a cylindrical mould (but it did not change its plate-like shape). This process allowed us to align the crystal with the applied magnetic field (minor misalignments, ~ 5 degrees, may occur).

Pulsed magnetic field measurements of 1.5 kT s^{-1} were performed at the Pulsed Fields facility of the Katholic University of Leuven, Belgium. A coil with an inductance of $650 \mu\text{H}$ was used to generate magnetic field pulse. This facility allowed us to go up to 70 T with pulse duration of 20 ms by discharging the capacitor bank through a specially designed magnet coil [20].

Magnetization reversal was detected using a compensated coil (see figure 1(a)), which was done by tuning the number of inner and outer windings. The compensated coil was sensitive to the signal coming from the sample and insensitive to the signal coming from the applied pulses. Low temperatures down to 0.6 K were achieved using a He^3 cryostat. The sample holder was made of non-metallic materials and was submerged in liquid He^3 during the measurements. The temperature was varied in the range between 0.6 and 4.1 K by pumping the helium with control through a needle valve.

The crystal was initially magnetized to negative saturation at a fix bath temperature. Then we swept the magnetic field at an ultra fast rate of 1.5 kT s^{-1} , so the Fe_8 crystal is in a non-equilibrium magnetization state, as shown in figure 1(b). We recorded the signals of the compensated coil during the complete field sweep. The magnitude of the peak voltage captured with the coils, V , is proportional to the magnetic flux variation, $\text{d}B/\text{d}t$, being therefore $V \propto \text{d}M/\text{d}t$ of the sample and indicating the spins reversal. Figure 1(c) shows an example of the signal recorded as a function of the applied magnetic field during a magnetization reversal.

3. Results

Figure 2 shows the temperature dependence of the magnetization reversal for a sweeping magnetic field of 1.5 kT s^{-1} . We first plotted eight representative curves at different temperatures in figure 2(a) and we fitted the curves with three Lorentzian peaks corresponding to the spin reversal at each resonance ($H_z = nH_R$ with $\mu_0 H_R \sim 0.26 \text{ T}$ and $n = 1, 2$, and 3 being the order of the resonance). Each curve corresponds to the reversal of the magnetization at a fixed bath temperature. A color-scale 2D plot shows in figure 2(b) all the measured curves for temperatures between 0.6 and 4.1 K . We can see both the variation in amplitude and position of the peaks. Depending on temperature, the resonant field $H_z = nH_R$ changes, from the third resonance at low temperatures $T < 1.2 \text{ K}$ to the first resonance at high temperatures $T > 2.4 \text{ K}$. Further, we observe that at certain temperatures, relaxation occurs throughout more than one resonance field.

We notice here that the magnetization-reversal peaks at 0.6 and 0.7 K show a different trend compared to the rest of the temperatures—and even the fits of the reversal curves seem to follow a different law. The resonances are observed at fields larger than the calculated values with equation (1) ($\mu_0 H_R \sim 0.26 \text{ T}$ instead of 0.225 T).

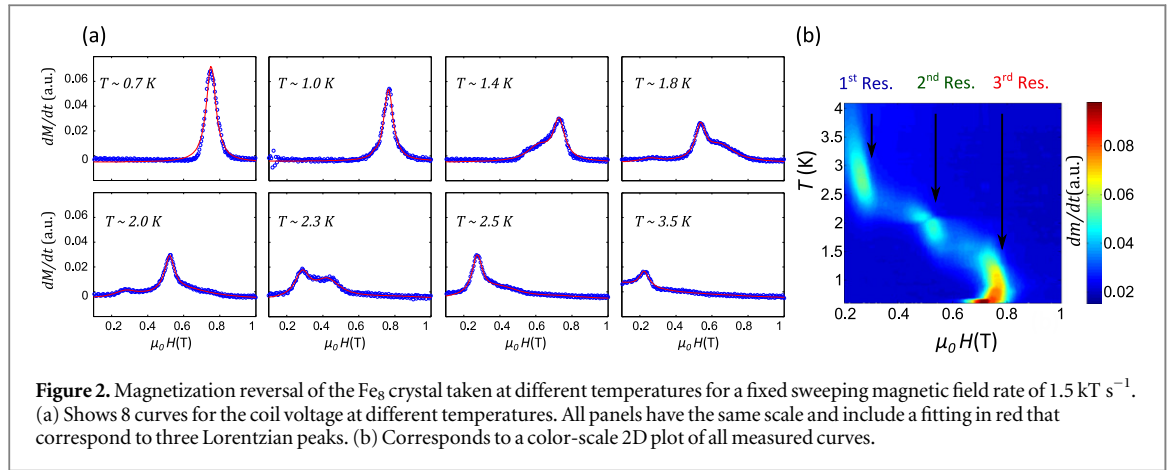


Figure 2. Magnetization reversal of the Fe_8 crystal taken at different temperatures for a fixed sweeping magnetic field rate of 1.5 kT s^{-1} . (a) Shows 8 curves for the coil voltage at different temperatures. All panels have the same scale and include a fitting in red that correspond to three Lorentzian peaks. (b) Corresponds to a color-scale 2D plot of all measured curves.

Previous experiments using high sweep rates showed similar field shifts [21]. We cannot attribute this large variation to a misalignment. We will discuss this effects in more detail later on.

As we increase the temperature we see how the reversal of the magnetization shifts towards lower magnetic fields while staying at the same resonant field. Peak position moves first within one resonance field and then jumps to the next smaller resonance field where it continues shifting with the same trend. The jump from the third dominant resonance field to the second one takes place around $T \sim 1.6 \text{ K}$ whereas the drop from the second to the first occurs around $T \sim 2.2 \text{ K}$. Detailed plots with peak analysis of the Lorentzian fits are displayed in figure 3. Panel (a) of figure 3 shows the resonance field values of the reversing magnetization peaks as a function of the temperature, panel (b) shows peak amplitude, and the panel (c), peak width. Peak amplitudes and peak widths have a negative correlation; when one increases the other one decreases and vice versa. This observation agrees well with having a total magnetization variation being roughly constant—especially when the reversal is dominated by a single peak. However, when we plotted the overall area under the coil's voltage (see, panel (d) of figure 3) we see that it slowly decreases with temperature suggesting the overall magnetization variation lowers with temperature. The initial magnetization state of our system once we begin measuring magnetization reversal (our measurements began at $\mu_0 H = 0.1 \text{ T}$) might be different because some spins might have already reversed at the zero field resonance [15].

4. Discussion

There are two remarkable features in our experiments compared to the usual spin dynamics observed at slow field sweep rates. First, a significant variation of the resonance field within the same-resonance level as a function of temperature, and second, a fast reversal of the magnetization occurring at low order resonances.

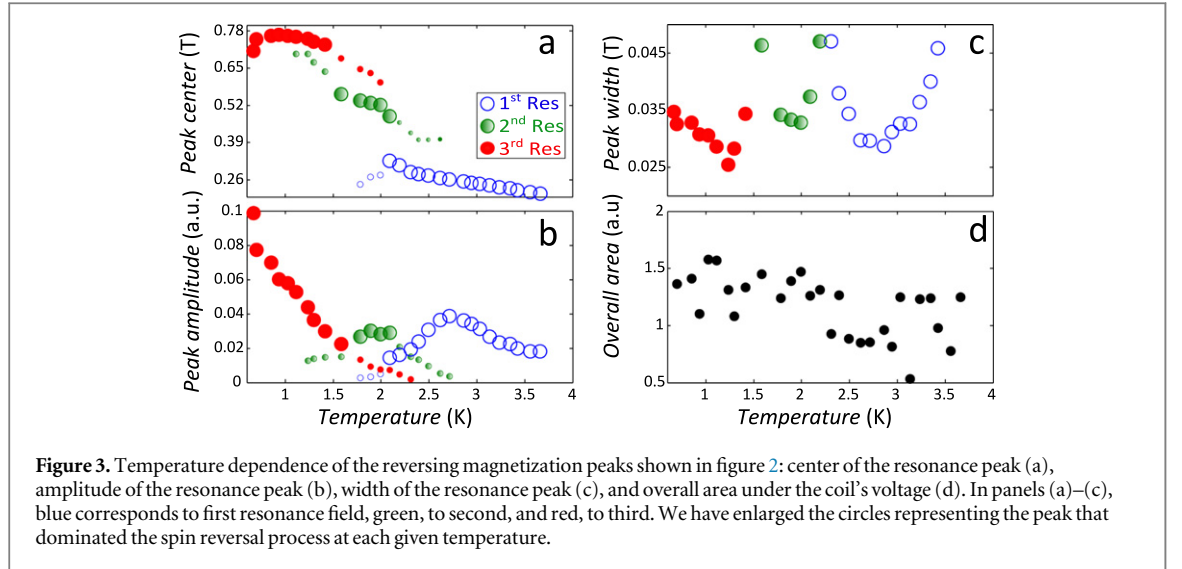
At a first order spin levels in Fe_8 molecule resonate all at the same applied field and variations in temperature (i.e., variations in the spin-level population) does not vary the effective resonance field. The presence of higher-order terms (\mathcal{H}_{ho}) in the Hamiltonian, such as S_z^4 would cause that resonance levels have a non-even spacing and therefore tunneling at different levels would occur at slightly different fields (this is the case of the Mn_{12} molecule magnet [22]). However, a fourth-order term of at least one order of magnitude higher than the measured one [18] would be necessary to explain the observed shifting of the resonance fields.

We believe that both the observation of the resonance shifting and the fast spin-dynamics observed are due to the emergence of a thermal instability—that may result in a magnetic deflagration—related with the self-heating process due to fast reversal of magnetization. In the following paragraphs we compare slow relaxation against ignition of a magnetic deflagration due to a thermal instability.

Magnetic deflagration has been extensively studied in $\text{Mn}_{12}\text{-ac}$ [19, 23–25], including experiments at high-field sweep rates [26]. In a magnetic relaxation process there is a competition between heat produced by the reversing spins and heat diffused throughout the sample and the bath; if the diffusion cannot compensate the reaction term, the process becomes unstable and some magnetic materials [23, 27, 28] experience a transition between thermal relaxation and a fast-propagating spin-reversal process named magnetic deflagration [19]. Additionally Leviant *et al* [29] have recently shown that magnetic deflagration might also occur in Fe_8 crystals.

The dynamics of the magnetization system when relaxing could be described by the following equations

$$\dot{M} = -\Gamma(M - M_{\text{eq}}), \quad \dot{T} = \dot{M}\Delta E/C + \nabla \cdot \kappa \nabla T, \quad (2)$$



where $\Gamma = \tau^{-1}$ is the relaxation rate, M_{eq} is the equilibrium magnetization, ΔE the energy released, C the heat capacity and κ the thermal diffusivity. The condition for the system to lose stability is given by

$$\dot{T} = 0, \quad \partial \dot{T} / \partial T = 0. \quad (3)$$

According to the theory of magnetic deflagration [25, 30], the threshold for ignition of the deflagration process is achieved when the rate, Γ , of the transition out of the metastable well exceeds a critical value (this follows from equations (2) and (3))

$$\Gamma_c = \frac{8\kappa k_B T_0^2}{U_{\text{eff}} \Delta E n_i l^2}, \quad (4)$$

where $U_{\text{eff}}(H)$ and $\Delta E(H)$ are the field-dependent effective energy barrier and the energy difference between spin-up and spin-down ground states, respectively, l is a characteristic length of the order the smallest dimension of the sample, n_i is the initial population in the metastable well and T_0 is the bath temperature. Once the relaxation rate is larger than the critical value given in equation (4) it means that the heat produced by the reversing spins cannot be compensated by the diffusion and therefore the sample's temperature will increase resulting in a faster spin relaxation and a faster heat production. Eventually, the magnetization reverses completely and the sample's temperature cools down to the bath temperature.

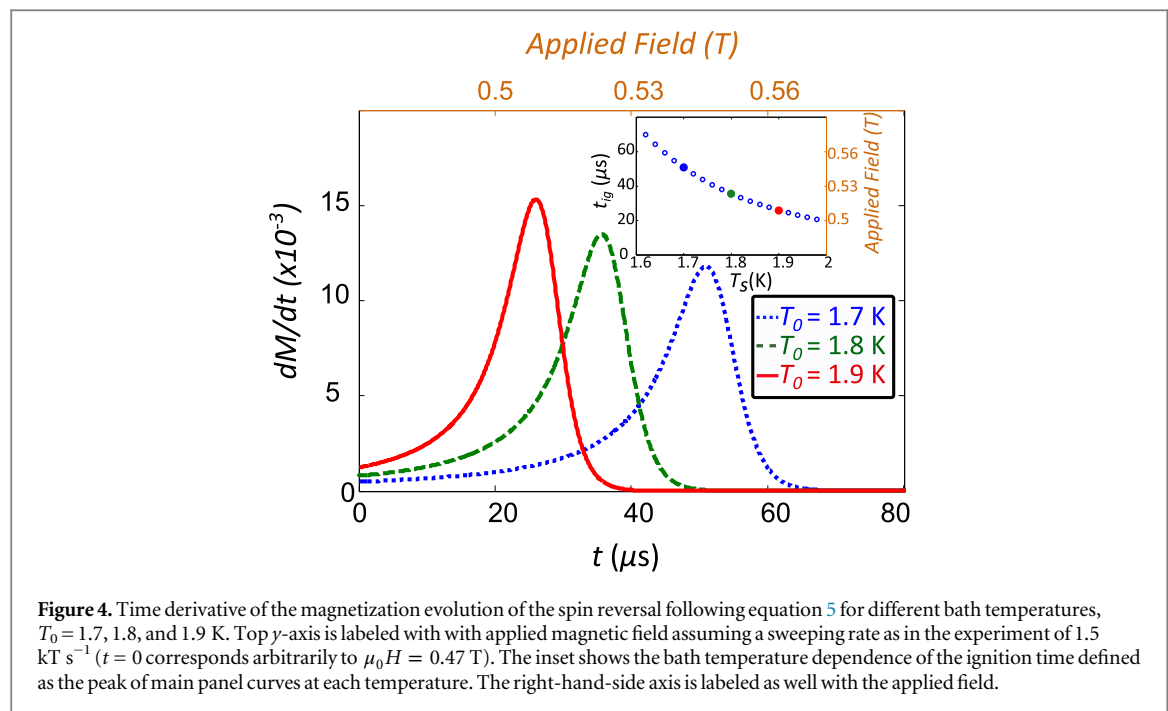
Considering our experimental conditions we now proceed to estimate the threshold for the relaxation rate that would induce a magnetic deflagration (see equation 4). For a temperature of $T = 2$ K at the second resonance, with an estimated effective barrier, U_{eff} , of about 15 K (see the appendix), we obtain $\Gamma_c = 20$ Hz – 2 kHz depending on the value for the thermal diffusivity, ($\kappa = 10^{-6} - 10^{-4} \text{ m}^2 \text{ s}^{-1}$). However, in our experiment the magnetization reverses in less than 100 μs —that corresponds to a spin relaxation rate of $\Gamma \sim 10$ kHz. This indicates that the spin reversal in our system should be driven by the emergence of a thermal instability. Once the relaxation rate reaches the critical value, a deflagration would develop and the sample temperature will increase leading to an increase of the relaxation rate [19, 31]. Note that reported experiments in Fe₈ at much slower field sweep rates [15] showed lower relaxation rates, on the order of a second, and thus the deflagration condition was never fulfilled.

The ignition of a thermal instability is sensitive to the bath temperature (see, equation (4)) [32]. Once the system reaches the instability—and spins begin to reverse and temperature, to increase—there is still a waiting time until the deflagration front forms or, in other words, until the temperature rises enough to allow a spin-reversal at the timescale of our sweeping field. This waiting period is sometimes called *ignition time* [24].

Next we compute the ignition times as a function of the bath temperature for the second resonance where we consider an effective barrier of 15 K (we describe the case in the appendix where we calculated the effective barrier, see figure 5(b)). Let us take here the magnetization and temperature evolution in a nucleation volume, independent of coordinates, as described in equation (2)

$$\dot{M} = -\Gamma(M - M_{\text{eq}}), \quad \dot{T} = \dot{M} \Delta E / C + \frac{\kappa}{2l^2}(T - T_0), \quad (5)$$

where T_0 is the bath temperature and $2l$ is the characteristic size of the nucleation volume and is bounded by the smallest sample dimension (0.15 mm). The diffusive term in equation (5) is linear with temperature while the



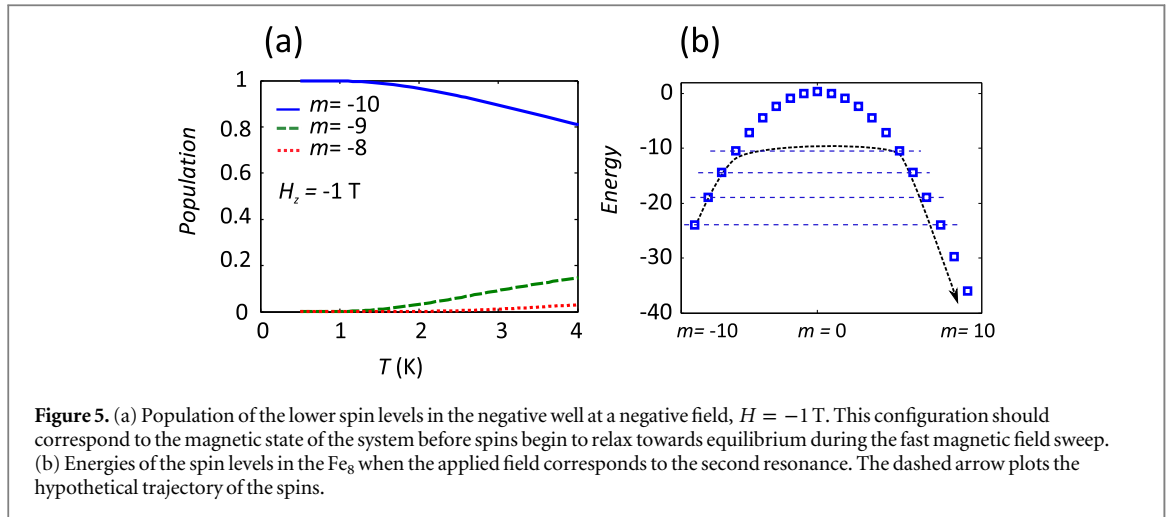
reaction term increases exponentially with temperature ($m \propto \exp[-U/(k_B T)]$). The competition between this two terms sets the critical value, T_c , for the thermal instability.

We have computed magnetization and temperature evolutions from equation (5) at different bath temperatures. Figure 4 shows representative time derivatives of the magnetization curves computed at bath temperatures $T_0 = 1.7, 1.8$, and 1.9 K (cases where spin relaxation occurs at the second resonance), using a heat capacity close to the measured value $C/(k_B N_A) \sim 1$ (i.e., $C = 8.3 \text{ J (mol}^{-1} \text{ K}^{-1})$) [33] and a thermal diffusivity of $\kappa = 10^{-5} \text{ m}^2 \text{ s}^{-1}$ [34]. We fitted the experimental data by varying the attempt frequency and obtained a value of $\Gamma_0 = 1.6 \times 10^7 \text{ Hz}$, which agrees well with reported values in the literature [3] for molecule magnets. However, we notice that these computations are very sensible to the parameters Γ_0 and C that determine the speed of temperature change in equation (5).

The ignition time depends strongly on the bath temperature and in our experiment, where field varies at 1.5 kT s^{-1} , this results in a shift of the field where the spin reversal is detected. Concretely, as the temperature increases the reversal of magnetization occurs earlier because the thermal instability develops faster. We labeled the top y-axis of the figure 4 main's panel and the right-hand-side axis of its inset with the applied field accounting for the used sweeping rate of 1.5 kT s^{-1} (we took $t = 0$ for the field $\mu_0 H_z = 0.47 \text{ T}$). We notice that the computed curves have a width of $20\text{--}30 \mu\text{s}$ that corresponds to $30\text{--}45 \text{ mT}$, which agree well with experimental curves (see figure 3). Additionally, the ignition time also gives a consistent explanation for the observed unexpected shift of the measured resonance-field towards higher values, rather than a large misalignment.

Finally, we notice that the lowest measured temperatures ($0.6\text{--}0.8 \text{ K}$) show a different trend in the magnetization reversal when comparing it with all other higher temperatures. We were not able to measure other curves in this regime and with the present data we are unable to describe the origin of this anomaly. However, given the high field-sweep rate used in our study, we cannot rule out the collective relaxation (superradiance) as a possible mechanism for the fast spin reversal observed in our measurements. An evidence for such scenario comes from the field-sweep-rate dependence of the low-temperature magnetization reversal shown by M Jordi *et al* [21]. Superradiance may also be relevant to the deflagration scenario described above as a mechanism responsible for the enhancement of the under barrier transitions (conventional tunneling mechanisms may quench because of the high field-sweep rate). Another possible scenario in this context might be the Fe_8 molecule magnet undergoing a phase transition to a ferromagnetic state at $T = 0.6 \text{ K}$ [35]. This would change the interactions among molecules and consequently the dynamics of the whole system.

In conclusion we have measured spin reversal through quantum tunneling at temperatures up to $\sim 4 \text{ K}$ in the Fe_8 molecular magnet using ultra fast pulsed magnetic fields of 1.5 kT s^{-1} . Measuring with high-fields sweep rates allowed us the observation of quantum spin-dynamic effects with spin populations not restricted to the ground states. Our experiments show a temperature dependence that suggests the spin reversal undergoing a thermal instability that probably causes a magnetic deflagration. Molecular magnet Fe_8 has a biaxial anisotropy



that brings interesting properties related to quantum tunneling of the magnetization. Thus observation of magnetic deflagration in the Fe_8 molecular magnet may open ahead new possibilities such as the observation of dipolar-field mediated deflagration [36]. At the lowest temperatures, the reversal process might develop concomitantly with collective electromagnetic or phonon emission. Establishing the precise mechanism of those complex relaxation processes be the subject for further experimental and theoretical studies.

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Appendix

To understand the temperature dependence of the fast spin reversal and eventually consider an effective barrier, U_{eff} , at a given applied field we need to account for all transition probabilities among levels—thermal activation, decay, and tunneling—and all level populations. The population variation of levels as a function of applied field, H_z , and temperature, T , are well known. Figure 5(a) shows the population of the lower levels for a large negative field (-1 T). This population configuration is similar to the population configuration right before the spins begin to relax towards equilibrium when H is swept from negative to positive values in our experiments. A longitudinal field H_z can drive the crystal in and out of tunneling resonances, reducing effectively the barrier separating spin-up and spin-down states (see, figure 5(b)).

The computation of the splitting $\Delta_{m,m'}$ of the (m, m') -resonance, in a given sample is not easy because the crystal is a macroscopic object that has demagnetizing fields both h_x and h_z that depend on the crystal shape and the overall tunneling is very sensitive to the transverse applied fields and it has a parity effect [5]. For instance, in absence of applied h_x , a single molecule will only have tunneling at the even resonances ($n = 0, 2, 4 \dots$). However, experimentally it is observed that both even and odd resonances show quantum tunneling. The presence of transverse fields varies critically, mixing among spin states, and allows tunneling between odd resonance levels [5]. In our experiments we have mentioned that there might be a misalignment, mainly because the crystallographic structure of Fe_8 . For instance, a small misalignment of 7 degrees would produce a transverse magnetic field of 0.1 T at the third resonance.

In order to estimate values for the level splittings we calculate a simple case where there are no transverse fields (i.e., we have to compute even resonances). Using perturbation theory [1] the values for the rate of quantum transition, $\Gamma_{m,m'} = \frac{\Delta_{m,m'}^2}{2\Gamma_0 \hbar^2}$ with $\Gamma_0^{-1} = \tau \sim 1 \times 10^{-7}$ s, being the characteristic lifetime of excited levels, in Hz for the second resonance ($H_z = 2H_R$)

$$\begin{aligned}\Gamma_{-10,8} &= 1.1 \times 10^{-9}, \Gamma_{-9,7} = 9.5 \times 10^{-5}, \\ \Gamma_{-8,6} &= 1.9 \times 10^0, \Gamma_{-7,5} = 1.3 \times 10^4, \\ \Gamma_{-6,4} &= 2.9 \times 10^7.\end{aligned}$$

Once $\Gamma_{mm'}$ exceeds the thermally activated relaxation rate from the m th level, the barrier is effectively reduced due to underbarrier tunneling from the m th level (see, figure 5(b)). Considering that the reversal of magnetization in our experiments occurs at about 10 KHz, the first quantum transition that would allow some variation of the magnetization is $\Delta_{-7,5}$ (and $\Delta_{-6,4}$ for a complete one), indicating that the effective barrier would be reduced to about 15 K.

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